Test by NMR of the Phase Coherence of Electromagnetically Induced Transparency

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Electromagnetically induced transparency is an effect observed in atomic systems, originating from quantum interference, in which electromagnetic transitions to and from a certain quantum state become suppressed. This dark state is also characterized by a quantum phase, relative to other states, which theoretically should stop evolving, but remain phase coherent, during transparency. We test this theoretical prediction using techniques developed for liquid-state nuclear magnetic resonance quantum computation, applied to a spin-7/2 nuclear spin system. A sequence of quantum operations is applied to create the dark state, and during transparency its phase evolution is measured relative to a reference state using Ramsey interferometry. Experimental measurements of the fringe visibility are in excellent agreement with theoretical expectations, taking into account measured decoherence rates.

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Many interesting macroscopically observable effects are known to arise from quantum interference, even in small, nearly perfectly closed quantum systems. Electromagnetically induced transparency (EIT) is a classic example of these effects at work in a three-level quantum system. First observed in atomic vapors [1], EIT is now employed as a general technique in a wide variety of physical systems, and may become an integral method for storage of quantum information [2–6] and probing decoherence [7].

Optical experiments in atomic vapors have produced measurement results consistent with theoretical prediction for effects such as EIT. However, certain quantum aspects of the theory have remained untested, because full control over the quantum systems has not been exercised. In particular, the canonical experimental test of EIT shows suppression of absorption of light at some frequency ω_{probe}, when light at another frequency ω_{control} is simultaneously applied. Quantum mechanically, this effect is understood as being due to the control field creating a dark state $|D\rangle$, turning off the interaction of the system with the probe field. From the theory, it follows that this dark state actually does not evolve at all during application of the control field; furthermore, since it is a quantum state, it can carry a phase relative to other states, which should remain unchanged while the system is transparent.

Knowing whether the dark state in EIT remains phase coherent during transparency is crucial to being able to use EIT as a means to faithfully store quantum information. This is because when several coupled dark states store the quantum information, then a phase evolution leads to errors, and, hence, decoherence. while indirect evidence for such phase coherence can possibly be derived from various atomic experiments, more interesting, perhaps, is that a direct test of this phase coherence can indeed be carried out, and that this test provides a quantitative measure of decoherence mechanisms at work during transparency.

The dark state phase coherence can be measured in the following manner: Introduce a fourth level $|R\rangle$ to the system (outside of the Hilbert space of the original three-level system) to act as a phase reference, then perform a Ramsey interference measurement [8,9]. The visibility of interference fringes created by Ramsey pulses before and after the control field would then provide a measure of the coherence of the dark state during transparency. In theory, the measurement should result in high fringe visibilities; otherwise, the dark state would have to be undergoing some unknown phase evolution. In this Letter, we shall be focused on the relative phase between the dark state $|D\rangle$ and the reference state $|R\rangle$; where there is no ambiguity, for simplicity this phase may be referred to as the phase of the dark state.

Here, following in the footsteps of prior demonstrations of atomic physics phenomena using nuclear magnetic resonance (NMR) [10], we implement this test of dark state phase coherence, using three levels of a nuclear spin-7/2 system as an analogue for the canonical three-level atomic system. Pulsed NMR methods and initial state preparation techniques developed for NMR quantum computation [11,12] are used to initialize the system, construct the dark state, and implement the Ramsey interference measurement. Just as in the atomic vapor case, electromagnetic radiation is used as the probe and control fields, but in contrast with the atomic case, measurement are made not on the transmitted field intensities, but rather on the spin system itself, by measuring the magnetization in a phase sensitive manner traditional to NMR. This permits a direct measurement of the quantum state.

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The EIT effect in a three-level quantum system can be understood as follows. Consider a Λ-like atomic configuration [2,3], and denote the energy eigenstates as $|1\rangle$, $|2\rangle$, and $|3\rangle$. The energy difference between the $|1\rangle$ and $|2\rangle$ and the $|2\rangle$ and $|3\rangle$ states corresponds to $\omega_{\rm probe}$ and $\omega_{\rm control}$, respectively. Figure 1 sketches the Λ-like system and its translation into a three-level NMR spin system.

Two electromagnetic fields are applied simultaneously to the system: The probe field at frequency $\omega_{\rm probe}$ of strength a and the control field at frequency $\omega_{\rm control}$ of strength b. The Hamiltonian describing the system-light interaction in the rotating wave approximation can be written (in the $|1\rangle$, $|2\rangle$, and $|3\rangle$ basis) as

$$H_{\rm EIT} = a|1\rangle\langle 2| + b|2\rangle\langle 3| + \text{c.c.} \qquad (1)$$

The numbers a and b can in general be complex, but we choose them to be real in our experiments. Two interesting regimes of the EIT effect exist, and we treat each of these separately below.

(1) EIT in the strong control field regime.—Here, the strength of the control field is much larger than the probe field, $b \ll a$, such that the dark state is $|D\rangle = |1\rangle$. In this regime, the time evolution of the system is given by

$$e^{-iH_{\rm EIT}t} \approx \begin{bmatrix} 1 & i\frac{a}{b}\sin(bt) & \frac{a}{b}[\cos(bt)-1] \\ i\frac{a}{b}\sin(bt) & \cos(bt) & i\sin(bt) \\ \frac{a}{b}[\cos(bt)-1] & i\sin(bt) & \cos(bt) \end{bmatrix}, \qquad (2)$$

to first order in $\frac{a}{b}$. From Eq. (2), we find that the matrix element connecting states $|1\rangle$ and $|2\rangle$ is zero when $b \gg a$. This means that the spin system becomes transparent to the $|1\rangle \rightarrow |2\rangle$ transition frequency ($\omega_{\rm probe}$) in the strong control field limit.

(2) Coherent dark state EIT.—Here, the control and probe fields are of a fixed ratio such that the dark state is the superposition state $|D\rangle = (1/\sqrt{a^2+b^2})(b|1\rangle - a|3\rangle)$. This follows simply from recognizing that $|D\rangle$ is an eigenstate of the EIT Hamiltonian, Eq. (1), with eigenvalue zero. When the spin system is in the zero eigenvalue state, it does not evolve under the EIT Hamiltonian. Hence, the EIT system becomes transparent to both the probe and control frequencies simultaneously [13]. For example, when $a = b$, the dark state is $(|1\rangle - |3\rangle)/\sqrt{2}$.

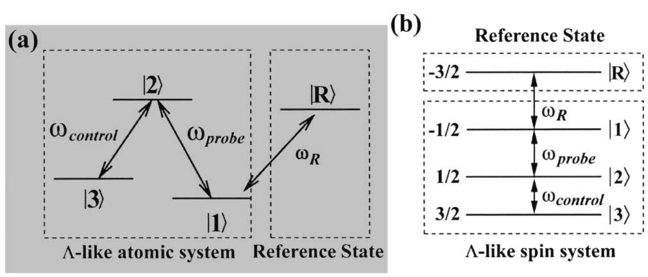

FIG. 1. (a) Energy level diagram of Λ-like spin states of a three-level system coupled to an additional reference level $|R\rangle$. (b) Schematic energy level diagram for the $I_z = (-3/2, -1/2, 1/2, 3/2)$ levels of a higher-order spin system with quadrupole coupling in the presence of an external magnetic field.

Traditionally, dark states in atomic systems are prepared by slowly ramping the probe field in the presence of a control field to adiabatically evolve the state from the initial ground state $|1\rangle$ to the stationary state of the EIT Hamiltonian [2–4]. Here, we use a different approach for creating dark states for arbitrary a and b based on quantum computing techniques for higher-order spin systems [14].

Starting with the $|1\rangle$ state, we apply a σ_x rotation in the $\{|1\rangle, |2\rangle\}$ subspace for an appropriate duration to obtain $(b|1\rangle + a|2\rangle)/\sqrt{|a|^2+|b|^2}$. We then apply a NOT gate on the $\{|2\rangle, |3\rangle\}$ subspace, with an overall π phase, giving us the appropriate dark state $(b|1\rangle + a|3\rangle)/\sqrt{|a|^2+|b|^2}$. The required operations are achieved by using single "qubit" gates previously developed [14].

The phase of the dark state $|D\rangle$ (relative to the reference state $|R\rangle$) should, in theory, remain unchanged during the EIT evolution period [4]. In an NMR system, this can be tested by coupling the dark state to an additional level $|R\rangle$ [or example, the $I_z = -3/2$ spin state as shown in Fig. 1(b)] to allow measurement of the phase of the dark state through Ramsey interferometry. This interference experiment is a direct measure of the phase coherence of $|D\rangle$, because the EIT Hamiltonian does not act on $|R\rangle$, and the experiment is an indirect measure of the transparency of the system to the probe field. Below, we shall employ $H_{\rm EIT}$ as defined in Eq. (1), but extended trivially to act as an identity on $|R\rangle$.

The basic idea of Ramsey interferometry is to measure phase evolution by creating a superposition state, allowing evolution to occur, then undoing the superposition and measuring the probability of observing the final state to be the same as the initial state. This is implemented to measure the phase evolution of $|D\rangle$, in the following manner, using a fourth state $|R\rangle$ as a phase reference for $|D\rangle$. We perform the sequence of four operations:

$$U_{\rm Ramsey} = H_{(R,D)} e^{-iH_{\rm EIT}t} e^{i\theta\sigma_z} H_{(R,D)}, \qquad (3)$$

where $H_{(R,D)}$ is a Hadamard operation and σ_z is a Pauli operation in the two-dimensional space defined by $\{|R\rangle, |D\rangle\}$.

Let $p = \langle D|e^{-iH_{\rm EIT}t}|D\rangle$ be the probability amplitude to stay in the dark state after evolving under the EIT Hamiltonian for time t. If the initial state of the system is $|R\rangle$, then the probability of obtaining $|R\rangle$ as the final state after $U_{\rm Ramsey}$ is

$$I(\theta) = |\langle R|U_{\rm Ramsey}|R\rangle|^2 = \frac{1 + p^2 + 2p\cos\theta}{4}. \qquad (4)$$

The final measurement of the NMR system is proportional to $I(\theta)$, and gives the visibility

$$V(a, b) = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}}, \qquad (5)$$

as a function of a and b, where the maximum and minimum of I are taken over θ. Note that H_{EIT} is applied for a constant time duration, varying a/b. p is ideally expected to be equal to 1, if the dark state remains coherent during the EIT evolution period.

Experiments were performed to test the dark state phase coherence using this procedure at MIT using a custom-modified Varian Unity Inova 11.7 T spectrometer with a Varian H-X probe. We used an $I = 7/2$ cesium nucleus oriented in a nematic liquid crystal phase as our higher-order spin system. The sample was prepared by mixing 50% by weight each of cesium pentadecafluor-ooctanoate and D_2O [15]. The spin of cesium has a Larmor frequency of 65 MHz at the given field strength. To form a spin-3/2 system, we use the central four energy levels $I_z = -3/2, -1/2, 1/2, 3/2$ because they have the longest coherence times [14] (see Figs. 1 and 2).

The longitudinal coherence times of the transitions were measured using the inversion recovery method, and were experimentally found to be $T_{1,\mathrm{control}} =$ 60–70 ms, $T_{1,\omega_R} =$ 60–70 ms, and $T_{1,\omega_{\mathrm{probe}}} = 120$ ms. The transverse coherence times, T_2, were obtained via spin echo techniques and are roughly equal to the measured T_1 times. The experiments were performed at a temperature of 27 °C, which gives the best trade-off between line shaped, decoherence times, and energy level splittings. The energy level splitting at this temperature is about 7.5 kHz [16].

At room temperature, the initial state is highly mixed [17]. Experiments in the strong control field limit were performed directly on this thermal state. This is possible because the final observable for the signal intensity is the $|R\rangle\langle 1|$ quantum coherence which undergoes the same evolution independent of whether a mixed or effective pure state is used. The coherent dark state EIT experiments were performed using the method of temporal labeling [18], which employs a sum over two experiments to isolate the signal coming from the desired initial state $|R\rangle$.

The required quantum gates for realizing dark states and the necessary unitary transforms for Ramsey interferometry as well as the readout of spin states were achieved using transition selective Gaussian shaped pulses with a duration of 620 μs. The pulse length was designed to be as short as possible without significantly exciting the neighboring transitions.

The pulse sequence for $H_{(R,D)}$ in the strong control field limit is given by $Y_R^1 Z_R^2$, written with time going from left to right. The subscript denotes the transition on which the pulse was applied (see Fig. 1), the superscript denotes the rotation angle in units of $\pi/2$, and X, Y, and Z denote the type of rotation (σ_x, σ_y, and σ_z, respectively). The $\hat{z}$ rotation for the Ramsey interference, Z_R^θ, can be implemented by shifting the phase of subsequent pulses.

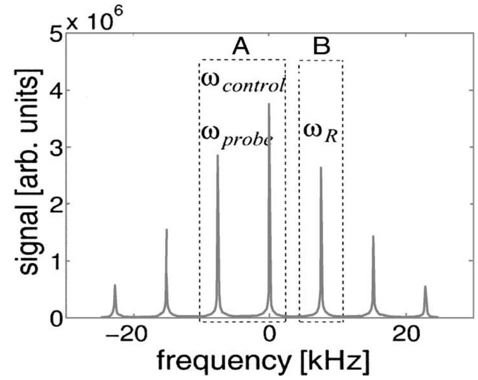

FIG. 2. Thermal NMR spectrum of ^{133}Cs in a nematic liquid crystal, with frequency given relative to 65 MHz. Seven distinct transitions are clearly seen due to the quadrupolar interaction between the external electric field gradient and the nucleus.

The pulse sequence for $H_{(R,D)}$ in the coherent dark state limit, when $a = b$, is given by $X_C^2 X_P^1 Y_R^{-1} Z_C^2 Z_P^4 Z_R^2 X_P^1 X_C^2 Z_R^3 Z_P^2 Z_C^1$. We use a simplified version of $H_{(R,D)}$ to create the dark state $|D\rangle$, using the pulse sequence $Y_R^1 X_P^2 X_C^2$ and starting with the state $|1\rangle$. The $\hat{z}$ rotation for the interference is implemented via $Z_R^\theta Z_P^\theta$. The initial state is prepared by summing two experiments; the first is performed on the thermal population distribution while the second is performed after first applying the pulse sequence $X_C^2 X_P^2 X_C^2$. The effect of this summation was to prepare the initial effective pure state $|R\rangle$.

The EIT pulse is realized by turning on ω_{probe} and $\omega_{\mathrm{control}}$ at the same time using Gaussian shaped pulses for a duration of 6 ms. The length of the EIT pulse is made longer to allow for lower power settings. This permits sweeping through a wide range of control field intensities for characterizing the visibility without heating the sample or damaging the rf coil. The probe field power is set to a power corresponding to $3\pi/2$ pulses.

The experimental visibility measurements are obtained as follows. After each Ramsey interferometry period, we measure the probability of the state to be $|R\rangle\langle R|$ by applying a 90° readout pulse at frequency ω_R, resulting in an NMR signal proportional to the final population in the state $|R\rangle$. We then vary the applied phase θ [Eq. (4)], and record the obtained maximum and minimum NMR signal from which we calculate the visibility $V(a, b)$. This is all done for a fixed state $|D\rangle$, which is the true dark state only for specific values of a and b. The results, shown in Fig. 3, are given for $D = |1\rangle$ and $D = (|1\rangle - |3\rangle)/\sqrt{2}$, which are the dark states for $b \gg a$, and $a = b$, correspondingly.

We observe a maximum visibility of $\approx 75\%$ in the experiments. The deviation from the ideally expected curve (solid line in Fig. 3) is largely due to three effects: relaxation during the time of the experiments, pulse imperfections due to the rf inhomogeneity of the NMR probe, and transient Block-Siegert shifts [14,19]. Using

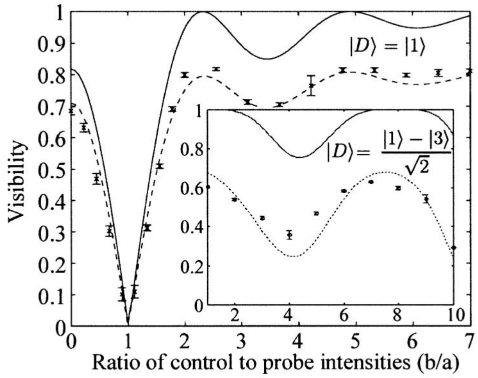

FIG. 3. Experimental results showing visibility as a function of b/a, for $|D\rangle = |1\rangle$, the strong control field dark state, and $|D\rangle = (|1\rangle - |3\rangle)/\sqrt{2}$, the coherent dark state (inset). $|1\rangle$ is the dark state for $b \gg a$, and thus the visibility is high for $b/a \gg 1$, but drops to zero for $b/a = 1$. The state $(|1\rangle - |3\rangle)/\sqrt{2}$ is a dark state for $b = a$, and thus the measured visibility is high at $b/a = 1$, but drops at other values. The solid line indicates the ideal theoretical expectation, the dashed line depicts numerically computed behavior taking into account decoherence effects and pulse imperfections, and the points indicate measured experimental results. The error bars indicate 95% confidence intervals estimated by repetition of identical experiments.

independent measurements of these parameters, and employing a relaxation model [20] generalized to higher-order spins using a Lindblad formulation [21,24], we obtain numerical simulation results (shown as dashed lines in Fig. 3) in good agreement with the experimental observations. The remaining sources of errors are estimated to be due to noise in the measurement signal and small fluctuations in the sample temperature.

These measurement results are in good agreement with theoretical expectations including relaxation and other nonidealities, and support the prediction that the EIT dark state remains phase coherent during transparency. Such coherence is necessary for storage of quantum information, in which EIT systems may play an important future role. This experiment also illustrates how the coherent control techniques employed in NMR quantum computation can be useful in testing basic physical phenomena; similar experiments could be performed on atomic systems using pulsed last excitation, implementing the same quantum logic gates and state preparation schemes. We believe this work is but a first step in such directions, and foresee a rich transfer of techniques from NMR to other physical systems, for complete experimental control of small quantum systems.

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